

AD 684 764

Reproduced by the  
CLEARINGHOUSE  
for Federal Scientific & Technical  
Information Springfield, Va. 22151

## CONTENTS

Abstract	ii
Problem Status	ii
Authorization	ii
INTRODUCTION	1
EXPERIMENTAL PROCEDURE	1
Phase 1	2
Phase 2	2
Phase 3	2
RESULTS AND DISCUSSION	4
Phase 1	4
Phase 2	6
Phase 3	6
Comment on PTS	9
SUMMARY	10
ACKNOWLEDGMENT	10
REFERENCES	11

## ABSTRACT

The ability of ten candidate fungicides, singly and in combination, to suppress fungal growth on a variety of organic coatings under natural, tropical conditions was investigated. After 1 year of exposure, substantial protection for the susceptible coatings was afforded by copper-8-quinolinate and binary mixtures of p-toluenesulfonamide with copper-8-quinolinate and phenylmercuric phthalate in fairly low concentration. At a higher concentration, p-toluenesulfonamide provided good protection for the susceptible coatings, while its N-ethyl derivative was less effective and its N-cyclohexyl derivative was almost inert. At an intermediate concentration, 2,3-dichloro-1,4-naphthoquinone and p-chlorophenoxyacetic acid provided effective protection for the susceptible coatings. None of the eight binary mixtures tested showed any evidence of synergism. The results confirm those obtained from earlier exposure programs, which showed that p-toluenesulfonamide is effective at higher concentrations in providing protection against fungal infestations. It was observed, however, that at very low levels this material may act as a growth promoter.

## PROBLEM STATUS

This is a final report on fungicidal coatings. Unless otherwise notified, this problem will be considered closed 30 days after the issuance of this report.

## AUTHORIZATION

NRL Problem C03-02  
Project RR007-08-44-5504

Manuscript submitted October 28, 1968.

## FUNGUS-INHIBITIVE COATINGS IN A JUNGLE ENVIRONMENT

F. T. Brannan,\* J. D. Bultman,† and J. M. Leonard†

### INTRODUCTION

For many years NRL has carried on an intermittent program pertaining to the study of fungal growth on organic coatings (1-5). Of particular interest had been the problem associated with the growth of fungus on coatings for electrical and electronic equipment. Failure of such equipment has occurred particularly under the adverse climatic conditions found in tropic regions. There was impairment of these coatings through the action of such organisms, but penetration of moisture into the insulating materials, with the attendant lowering of insulation resistance, was the usual cause of failure. The goal for the NRL program was to develop techniques which could be applied to the suppression of fungal growth on such equipment in tropic service, thereby prolonging its operational life.

This work has been extended to the development of fungus-resistant exterior coatings. Although the problem is essentially the same as that for electronic equipment — the incorporation of an effective fungicide into a vehicle with which it is compatible — consideration must also be taken of the effect of the fungicide on the aesthetic and other properties of these coatings. Chemicals which would cause discoloration in the finished coatings or chemicals which would inhibit their curing reactions or change their adhesive properties would not be suitable. Also, since exterior coatings may be exposed to large amounts of solar radiation, the photochemical stability of the fungicide must be good, particularly in the ultraviolet region of the spectrum. Any evaluation of a potential fungicide must also include an evaluation of these effects as well.

Some of the chemical agents used in the current exposure program have been used in a previous one (5) in which they were incorporated, singly and in binary mixtures, into a varnish coating. In the earlier study, these agents were found to impart significant protection to the varnish, and 12 combinations of the 15 binary mixtures employed showed a synergistic response. The current program serves, in part, as an extension of the earlier one, in that the behavior of some of these same agents in a variety of vehicles is being studied. Of particular interest are the coatings containing p-toluenesulfonamide and mixtures of this chemical with various others and the synergistic effects that may occur. Previous experiments by NRL (2, 3, 5) had shown this material to be highly effective. Until the work of the present report was completed, the writers had no evidence that any other investigator had tested p-toluenesulfonamide. Recently, Hoffmann, et al. (6), have followed the suggestion of the NRL work by trying the material in alkyd enamels.

### EXPERIMENTAL PROCEDURE

The present investigation was carried out in three phases over a period of 3 years; experimental details common to all three will be described first. Six coatings were chosen as carriers for the toxicants. Included in this group of coatings were a zinc-free exterior white paint, a glossy white enamel, a red "barn paint," a chlorinated-rubber

\*Present address — Army Tropic Test Center, Fort Clayton, C.Z.

†Present address — Marine Biology and Biochemistry Branch, Ocean Sciences Division, NRL.

paint, an alkyd varnish, and an alkyd-modified nitrocellulose lacquer. The first four coatings were prepared in the laboratory, and their compositions are given in Table 1. The remaining coatings were commercial preparations. The various toxicants were dispersed singly and in binary mixtures of equal concentration in the coatings by ball- and roller-mill techniques. In all cases the percentage of toxicant added was based on the amount of "nonpigment-solids" content of each vehicle. This base was used because it is believed that only the organic portion of the film required protection against fungus infestation and that comparisons between coatings would be more valid if the weighting effect of the pigment were eliminated.

For each of the experimental coatings three panels were prepared. The paint was applied to one face of a 1-in.-thick-by-1-ft-square yellow pine board. Each panel was given at least two coats, with thoroughly cleaned brushes being used for each formulation. Panels painted with unfortified coatings served as the controls. One day after painting, the test specimens were mounted, by a scheme of random numbers, in a vertical position on an exposure fence erected in the Panama jungle on the northern, or Caribbean, side of the Isthmus. The fence was covered by a narrow roof, which protected the panels from the washing action of direct rainfall and from direct solar radiation, while still allowing free air circulation. Figure 1 shows the exposure fence during installation of the test panels. The exposure site was chosen because of the excellent environmental factors conducive to fungal growth. The average annual rainfall at the site is about 130 in. and is concentrated in a rainy season of about an 8-month duration; the average temperature is about 80°F. The relative humidity is always high, and fungi flourish. The panels were examined at intervals, and fungal attack was rated on an arbitrary, visual scale of 0 to 5, the latter representing visible fungal attack over the entire painted area. Final examinations of the panels were performed in the laboratory at the site; interim examinations were made at the fence.

#### Phase 1

In the first phase of the investigation p-toluenesulfonamide (PTS), salicylanilide (S), and copper-8-quinolate (Cu-8) were used singly as the toxicants. In addition, PTS was combined in a 1:1 ratio with Cu-8, salicylanilide (S), pentachlorophenol (PENTA), and phenylmercuric phthalate (Ph-Hg). These chemicals were used in the earlier study (5). The experimental coatings contained the inhibitor at 0.5%, 1.0%, 2.0%, and 4.0% total concentration of toxicant.

#### Phase 2

In the second phase, PTS and two derivatives, N-cyclohexyl-p-toluenesulfonamide and N-ethyl-p-toluenesulfonamide, were used as the toxicants at 8% and 12% levels of concentration. Because the primary concern of this study was the toxicants, those coatings whose controls showed high natural resistance in Phase 1 were excluded from Phases 2 and 3, and only the susceptible ones — the exterior white, the gloss enamel, and the "barn paint" — were used.

#### Phase 3

In the third phase, toxicants which had shown promise in earlier work (5, 7) were used singly and in 1:1 binary mixtures. These chemicals were tetrachlorophenol (TETRA), 2,3-dichloro-1,4-naphthoquinone (DCN), and p-chlorophenoxyacetic acid (PCA). The same vehicles were used as in Phase 2, and the toxicants were present at 4% and 8% final concentration.

Table 1  
Coating Formulations

Components	Percent
Zinc-Free Exterior White Paint	
TiO <sub>2</sub>	11.1
Blanc fixe	29.7
Celite	11.1
Aluminum stearate	0.22
Soybean drying alkyd*	29.7
Raw linseed oil	3.7
Bodied linseed oil	3.7
Lead drier	0.291
Cobalt drier	0.22
Manganese drier	0.09
Mineral spirits	10.3
Red Barn Paint	
Linseed oil	59.7
Iron oxide	12.0
CaCO <sub>3</sub>	6.6
Celite	13.9
Lead drier	0.75
Manganese drier	0.23
Mineral spirits	7.7
Gloss White Enamel	
25% Phthalic soya alkyd†	50.6
TiO <sub>2</sub>	32.9
Cobalt drier	0.40
Manganese drier	0.005
Lead drier	0.89
Mineral spirits	15.2
Chlorinated-Rubber Paint	
Styrene-butadiene‡	19.5
TiO <sub>2</sub>	36.2
Chlorinated di- and polyphenyls§	1.96
Linseed oil	0.98
High-flash naphtha	20.7
Toluene	20.7

\*P296 resin, 70% solids.

†Glyptal 2466, 70% solids.

‡Phiolite S-5.

§Arochlor 1254.

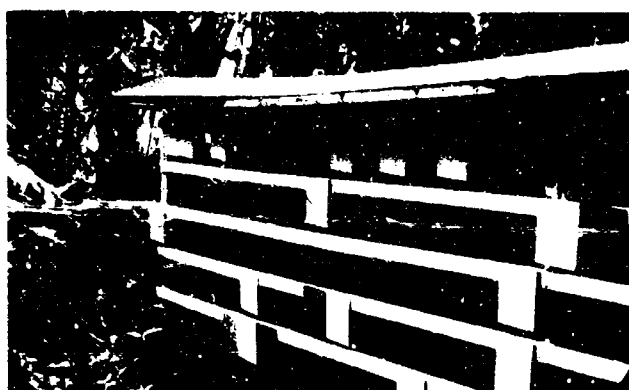


Fig. 1 - Exposure fence during installation of the test panels

## RESULTS AND DISCUSSION

### Phase 1

After 18 months of exposure of the test specimens of Phase 1, three coatings — the chlorinated-rubber paint, the clear lacquer, and the varnish — remained completely free of fungal growth, even in the absence of fungicide. The total resistance of these coatings to fungal attack under these conditions throughout the duration of the exposure period is interesting. Perhaps the chlorinated polyphenyls of the first vehicle are moderately toxic — the point should be examined further; since we did not manufacture the other two resistant vehicles, seeking explanations based on composition would be only speculative. Simple nutritional inertness of these coatings is not a sufficient explanation. All of them contain organic constituents which could serve as a carbon source and, thus should be susceptible to fungal attack. In addition, one could also expect surface deposits of organic debris sufficient to support visible growth; however, the smoothness of the panels and their vertical orientation on the exposure racks may have minimized such accumulation.

The exterior white, the enamel, and the barn paint, however, proved to be very susceptible to attack by fungus; none of the toxicants or combinations thereof were effective in protecting these coatings even at the highest (4%) level of concentration throughout the entire exposure period of 18 months. The exposure data for these coatings are presented in Table 2. Of these coatings, the barn paint suffered the heaviest attack, possibly because of its very high linseed oil content. Very little protective effect remained after 12 months of exposure, and this protection was provided mostly by salicylanilide at 2%, Cu-8 at 4%, and PTS + Cu-8 and PTS + Ph-Hg at 2% total concentration; anomalously, there was practically no protection of paint containing binary mixtures at 4% total concentration.

Table 2  
Ratings for the Susceptible Coatings of Phase 1

Fungicide	Percent	Extent of Fungus Growth on the Coatings by Panel (triplicate panels, arbitrary scale of 0 to 5)								
		Exterior White Paint			Enamel			Barn Paint		
		6 Months	12 Months	18 Months	6 Months	12 Months	18 Months	6 Months	12 Months	18 Months
Controls	0.0	1,4,-	4,5,5	5,5,5	3,3,-	5,3,5	5,5,5	4,5,5	4,5,5	5,5,5
PTS	0.5	4,4,-	4,5,5	5,5,5	4,-,-	4,5,5	5,5,5	5,5,5	5,5,5	5,5,5
PTS	1.0	4,5,-	5,5,4	5,5,5	3,3,3	4,5,4	5,5,5	5,5,4	5,5,5	5,5,5
PTS	2.0	5,5,-	5,5,5	5,5,5	4,3,-	5,5,5	5,5,5	4,-,-	5,3,3	5,5,5
PTS	4.0	5,5,5	5,5,5	5,5,5	2,4,-	5,3,5	5,5,5	4,0,-	5,5,4	5,5,5
PTS + S	0.5	5,4,-	5,5,5	5,5,5	4,3,3	4,5,5	5,5,5	5,3,0	5,5,5	5,5,5
PTS + S	1.0	5,1,-	5,5,3	5,5,5	4,3,-	5,4,5	5,5,5	5,4,5	5,5,5	5,5,5
PTS + S	2.0	4,3,3	4,4,3	5,5,5	3,2,-	5,5,4	5,5,5	5,4,4	5,5,3	5,5,5
PTS + S	4.0	2,2,0	4,2,1	4,3,5	3,1,2	2,2,2	5,4,5	5,3,4	5,3,5	5,5,5
PTS + Cu-8	0.5	3,2,5	5,4,5	5,5,5	3,4,4	4,5,5	5,5,5	4,4,5	5,4,5	5,5,5
PTS + Cu-8	1.0	3,4,2	5,5,3	5,5,5	4,5,-	5,5,4	5,5,5	5,5,-	5,5,5	5,5,5
PTS + Cu-8	2.0	1,1,0	1,1,1	5,5,5	3,3,5	4,5,5	5,5,5	3,0,0	0,1,1	5,5,5
PTS + Cu-8	4.0	1,0,-	1,0,1	5,5,3	2,1,1	2,5,1	5,5,5	4,4,-	5,5,4	5,5,5
PTS + PENTA	0.5	5,5,4	5,5,5	5,5,5	5,4,-	5,4,5	5,5,5	5,5,4	5,5,5	5,5,5
PTS + PENTA	1.0	4,5,3	4,5,5	5,5,5	4,3,5	4,4,5	5,5,5	3,-,-	5,5,5	5,5,5
PTS + PENTA	2.0	4,5,4	5,5,5	5,5,5	2,5,4	3,5,5	5,5,5	4,3,3	5,3,3	5,5,5
PTS + PENTA	4.0	1,4,-	5,3,3	5,5,5	4,1,-	3,3,5	4,5,5	4,5,2	5,5,5	5,5,5
PTS + Ph-Hg	0.5	3,2,1	4,4,4	5,5,5	1,1,-	0,5,2	5,5,5	5,2,3	5,5,3	5,5,5
PTS + Ph-Hg	1.0	1,0,4	2,5,1	5,5,5	1,0,0	2,2,1	5,4,3	5,3,-	5,3,5	5,5,5
PTS + Ph-Hg	2.0	1,0,0	1,0,0	5,2,5	1,0,1	2,3,4	5,5,5	4,0,0	4,1,4	5,3,5
PTS + Ph-Hg	4.0	0,0,0	0,0,0	1,1,3	1,1,0	3,4,4	5,5,5	4,0,-	5,5,3	5,5,5
S	0.5	4,2,1	3,4,5	5,5,5	1,1,0	5,4,4	4,5,5	4,5,-	5,5,5	5,5,5
S	1.0	1,4,0	2,4,2	5,5,5	1,4,-	5,4,5	5,5,5	3,-,-	3,5,5	5,5,5
S	2.0	3,3,0	5,1,5	5,5,5	3,1,0	4,1,2	5,3,5	2,3,3	5,2,1	5,5,5
S	4.0	- - -	- - -	- - -	1,1,-	2,5,1	5,5,4	5,5,-	5,4,5	5,5,5
Cu-8	0.5	- - -	- - -	- - -	- - -	- - -	- - -	4,5,-	3,5,5	5,5,5
Cu-8	1.0	- - -	- - -	- - -	3,1,0	4,2,1	5,5,3	3,0,0	4,3,4	5,5,5
Cu-8	2.0	0,0,0	0,0,0	4,4,5	1,2,0	0,5,4	5,5,5	1,3,-	5,3,3	5,4,5
Cu-8	4.0	0,0,-	0,5,0	5,5,5	0,0,-	4,5,0	5,5,4	- - -	0,5,2	5,5,5

At the end of 12 months of exposure, only the exterior white coating containing Cu-8, PTS + Cu-8, and PTS + Ph-Hg at 2% to 4% total concentration remained substantially free of fungus. While these particular treatments materially improved the performance of this coating for the first year of exposure, they failed almost completely during the final 6 months of the exposure period, the notable exception being paint containing PTS + Ph-Hg at 4% total concentration. Although these additives were providing some measure of control of fungal growth, there was no evidence of a synergistic response between PTS and either the Cu-8 or salicylanilide. For example, the exterior white paint containing PTS at 1% concentration after 6 months of exposure showed an average reading of 4.5, the binary mixture with salicylanilide at 1% total concentration of constituents rated 3, and salicylanilide at 0.5% concentration rated 2.3. It is clear that the improvement noted in the mixture PTS + S was generated by the addition of salicylanilide. An identical comparison can be made with the binary mixture, PTS + Cu-8; therefore, synergism in these cases must be ruled out.



Salicylanilide at 2% and 4% concentration improved the performance of the gloss white enamel for the first year of exposure, as did Cu-8. The binary mixture PTS + Ph-Hg was also effective, although, anomalously, the lower concentrations provided the best protection. Since Ph-Hg and PENTA were not used singly as toxicants, no information is available concerning a possible synergistic response with PTS. However, Ph-Hg has been reported as an ineffective fungicide in certain varnishes (5, 8), and in the present study the binary mixture of PTS + Ph-Hg proved to be one of the better combinations, certainly better than PTS alone. This behavior hints at a synergistic relationship between these compounds.

It was found that all of the white coatings containing Cu-8 developed a brownish-green discoloration after 6 months of exposure. This characteristic has been reported previously (7, 9). Also, at the low levels of concentration employed, PTS by itself appeared to stimulate growth somewhat more than the nontreated exterior white coating. This growth-stimulating effect of PTS, when present in low concentration, is not surprising; many toxicants behave in this manner when present in small amounts.

## Phase 2

After 16 months of exposure of the test specimens of Phase 2 (exposure data in Table 3), the exterior white and the enamel containing unsubstituted PTS at the 12% concentration were found to be well protected from fungus. Even at a concentration of 8%, PTS displayed good protective ability during the first 7 months at least. No discoloration of the paints was noted. The results obtained from the barn paint, however, are certainly anomalous. Possibly many of the anomalies associated with such systems are the product of microenvironmental factors which are not as yet well understood.

The effect of altering the chemical structure of the parent compound PTS is readily apparent when its performance is compared to that of the N-ethyl and the N-cyclohexyl derivatives. The ability of PTS to inhibit the growth of fungus decreased as the size of the substituent group increased. The N-ethyl derivative was generally not as effective as the parent compound over the entire exposure period at either concentration; however, it provided comparable protection through the first 7 months. The N-cyclohexyl derivative was completely ineffective. This correlation between size of the substituent group and decrease in the fungal resistance of the coating suggests that the mechanism involved may be one of steric hindrance rather than the removal of a necessary amine hydrogen by substitution.

These gross observations have been subjected to a variance analysis, presented in Table 4. The F ratios associated with the vehicle factor and its interactions suggest that all three vehicles are very similar. Concentration does not figure as a significant variable, probably because of the nullifying effect of the almost inert cyclohexyl compound.

## Phase 3

Data from 12 months of exposure of the specimens in Phase 3 (Table 5) revealed that TETRA at 4% concentration did not provide fungal protection for the exterior white paint or the barn paint and only light protection for the gloss white enamel; an increase in concentration to 8% provided some additional protection for the exterior white and barn paint only. The impotence of this compound is somewhat surprising, since it has displayed antifungal activity previously, though on a totally different substrate (10). It is also being used at 4% concentration, based on total solids, in paint applied to buildings in the Panama Canal Zone. The coating has proved satisfactory; however, the buildings involved are not in an actual jungle environment, and the stress on the coating is not as severe as that in

Table 3  
 Ratings for the Coatings Exposed in Phase 2

Fungicide	Percent	Extent of Fungus Growth on the Coatings by Panel (triplicate panels, arbitrary scale of 0 to 5)											
		Exterior White Paint			Enamel			Barn Paint					
		4 Months	7 Months	16 Months	4 Months	7 Months	16 Months	4 Months	7 Months	16 Months	4 Months	7 Months	16 Months
Controls	0.0	2,1,4	5,5,5	5,5,5	2,4,2	5,4,5	5,5,5	2,2,4	5,5,5	5,5,5	2,2,4	5,5,5	5,5,5
p-Toluenesulfonamide	8.0 12.0	0,0,0 0,0,0	1,0,0 1,1,0	4,2,3 4,3,1	1,0,0 1,0,0	1,1,2 2,1,1	4,5,4 2,4,2	2,1,1 0,1,0	1,1,1 0,1,1	3,3,1 5,5,5	2,1,1 0,1,0	1,1,1 0,1,1	3,3,1 5,5,5
N-Ethyl-p-toluene sulfonamide	8.0 12.0	1,2,0 0,0,1	2,2,3 1,1,1	5,4,5 5,2,4	1,1,0 0,1,1	1,1,1 1,2,2	4,5,5 1,2,2	0,2,3 1,0,1	5,2,5 1,1,2	5,5,5 1,5,5	0,2,3 1,0,1	5,2,5 1,1,2	5,5,5 1,5,5
N-Cyclohexyl-p-toluenesulfonamide	8.0 12.0	1,1,3 5,1,3	5,5,5 5,5,5	5,5,5 5,5,5	1,2,3 3,2,2	4,2,4 5,4,4	5,5,5 5,5,5	2,3,4 4,4,3	5,4,5 5,5,4	5,5,5 5,5,4	2,3,4 4,4,3	5,4,5 5,5,4	5,5,5 5,5,4

Table 4  
Variance Analysis of Phase-2 Observations

Factor	Sum of Squares	d. f.	Variance	F Ratio
Vehicle (V)	13.7	2	6.8	1.1
Time (T)	215.1	1	215.1	34.8*
Fungicide (F)	390.4	2	195.2	31.6*
Concentration (C)	11.1	1	11.1	1.8
Interactions				
V x T	0.4	2	0.2	0.03
V x F	29.6	4	7.4	1.2
V x C	0.7	2	0.35	0.06
T x F	53.7	2	26.9	4.4†
T x C	1.8	1	1.8	0.3
F x C	46.7	2	23.4	3.8†
Residual	95.0	16	6.18	—
Total	862.2	35	—	—

\*P 0.001. †P<0.05

Table 5  
Ratings for the Coatings Exposed in Phase 3

Fungicide	Percent	Extent of Fungus Growth on the Coatings by Panel (triplicate panels, arbitrary scale of 0 to 5)					
		Exterior White Paint		Enamel		Barn Paint	
		4 Months	12 Months	4 Months	12 Months	4 Months	12 Months
Controls	0.0	2,4,1	5,5,5	3,1,2	5,5,5	1,5,0	5,5,4
TETRA	4.0	5,3,1	5,4,5	0,5,0	3,5,3	0,5,0	5,5,5
TETRA	8.0	5,0,0	5,4,1	0,0,4	3,4,4	3,1,1	3,4,4
TETRA + DCN	4.0	0,1,1	4,4,4	4,1,2	4,4,5	0,2,0	5,4,3
TETRA + DCN	8.0	0,0,0	1,3,3	0,0,0	5,4,4	3,0,1	4,4,5
DCN	4.0	1,0,0	5,2,4	4,1,4	5,4,5	1,0,1	4,4,4
DCN	8.0	0,0,0	0,1,0	0,0,0	3,3,2	0,0,0	4,3,2
DCN + PCA	4.0	0,1,0	4,5,4	4,0,3	4,4,4	0,1,1	4,4,3
DCN + PCA	8.0	0,0,0	2,2,1	0,0,0	2,4,3	1,4,3	1,4,3
PCA	4.0	0,0,0	3,2,2	2,1,2	5,5,4	1,0,1	4,4,4
PCA	8.0	0,0,1	1,1,2	0,0,0	2,3,3	1,0,1	3,5,3
TETRA + PCA	4.0	0,1,0	3,4,2	2,0,0	4,4,4	1,3,0	5,5,4
TETRA + PCA	8.0	0,0,0	1,1,2	0,0,0	5,2,5	0,0,0	5,3,2

this exposure. Also a concentration figure based on total solids will provide a higher absolute amount of toxicant than the same figure based on nonpigment solids. As stated already, the latter basis was used here.

Those coatings containing DCN and PCA at 8% concentration generally received superior protection from fungal attack as compared to the controls. The mixture of these compounds, however, did not give a synergistic response; actually, antagonism is indicated. Also, nearly all of the toxicants and combinations thereof provided considerably more protection to the exterior white coating than to the gloss white enamel and the barn paint. This observation can also be applied to the results obtained for these susceptible coatings in the first two phases as well. A variance analysis of these data is presented in Table 6. In this case all the factors, including the first-order interactions, appear to be significant. The importance of the concentration variable as compared to the effect of fungicide is reminiscent of an earlier study (2). In both instances the quantity of fungicide is more important than the kind.

Table 6  
Variance Analysis of Phase-3 Observations

Factor	Sum of Squares	d. f.	Variance	F Ratio
Vehicle (V)	95.1	2	47.6	80.7
Time (T)	1073.4	1	1073.1	1815.
Fungicide (F)	93.8	5	18.8	31.7
Concentration (C)	138.9	1	138.9	234.
Interactions				
V x T	15.8	2	7.9	13.3
V x F	54.6	10	5.5	9.3
V x C	31.3	2	15.7	26.5
T x F	19.5	5	3.9	6.57
T x C	152.4	1	152.4	257.
F x C	19.3	5	3.9	6.57
Residual	21.9	37	0.59	—
Total	1716.0	71	—	—

#### Comments on PTS

The exposure results obtained sustain the authors' belief that PTS has real possibilities as a fungus inhibitor, even though the concentration of the toxicant required to achieve this protection appears to be rather high. Actually, the 12% concentration of PTS is not excessive when reckoned on the more orthodox basis of total solids. On such a basis it amounts to only 4.3%, 6.1%, and 7.8% for the exterior white, the enamel, and the barn paint, respectively. Similarly, the 8% concentration found to be effective in Phase 3 is proportionately reduced by the more orthodox method of computation. As stated in the Introduction, the recent work of Hoffmann, et al. (6), is the only other exposure experiments on PTS we know of. They find that it is ineffective on surfaces exposed to direct weathering and that the loss of effectiveness is due to leaching. It did work well indoors, however. None of our experiments contradict the Hoffmann findings. Our previous experiments (2, 3, 5) were conducted inside a jungle hut; the panels in the present experiment were well shielded from the elements by the roof over the test fence (Fig. 1) and the high density of jungle vegetation. The merit which the compound shows in the present outdoor experiments probably reflects the protection of the panels from rainfall.

## SUMMARY

1. Three organic coatings — a chlorinated-rubber paint, an alkyd-modified nitrocellulose lacquer, and a clear alkyd varnish — were not attacked by fungus after 18 months of exposure in a jungle, even in the absence of toxic agent.

2. Three other organic coatings — an exterior white paint, a gloss-white enamel, and a barn paint — were readily attacked in the absence of toxicants. Of the three susceptible coatings, the exterior white coating appeared to respond best to added toxicant; the barn paint was most heavily attacked.

3. These latter coatings, particularly the exterior white paint, were afforded some protection against fungal attack, for 1 year only, by copper-8-quinolate and by binary mixtures of p-toluenesulfonamide with copper-8-quinolate and phenylmercuric phthalate, respectively.

4. Salicylanilide at comparatively low concentration and in combination with p-toluenesulfonamide improved the performance of the glossy enamel for 1 year. The binary mixture also improved the performance of the exterior white paint for the same period.

5. p-Toluenesulfonamide at the maximum test concentration provided protection against fungus attack in the three susceptible coatings for 16 months; at an intermediate concentration it was less effective.

6. The N-ethyl derivative of p-toluenesulfonamide was less effective than the parent compound; the N-cyclohexyl derivative was completely ineffective.

7. 2,3-Dichloro-1,4-naphthoquinone and p-chlorophenoxyacetic acid at the intermediate concentration provided effective protection for the susceptible vehicles for a period of 1 year; tetrachlorophenol was almost completely ineffective.

8. No synergistic response was observed in any of the binary mixtures of the fungicides.

9. p-Toluenesulfonamide in low concentration appeared to promote fungal growth on the test surface, particularly on the exterior white coating.

## ACKNOWLEDGMENT

The authors thank their colleague, Mr. Gerald E. Rohl, for preparing the coatings used in this study.

## REFERENCES

1. Williams, K.G., and Lempke, C.T., "A Study of Navy Radio Receivers in a Tropical Climate," NRL Report 3935, Feb. 1952
2. Leonard, J.M., and Pitman, A.L., "Tropical Performance of Fungicidal Coatings," NRL Report 3781, Jan. 1951
3. Leonard, J.M., and Pitman, A.L., "Tropical Performance of Fungicidal Coatings - A Statistical Analysis," Ind. Eng. Chem. 43:2338 (1951)
4. Leonard, J.M., and Larson, H.L., "Fungicide Attenuation by Oils," NRL Report 4228, Oct. 1953
5. Leonard, J.M., and Bultman, J.D., "Tropical Performances of Fungicidal Coatings, Part II," NRL Report 4563, July 1955
6. Hoffmann, E., Saracz, A., and Barned, J.R., J. Oil Colour Chemists' Assoc. 50:516 (1967)
7. Vicklund, R.E., and Manowitz, M., Paint Varnish Prod. 41:8,26 (1951)
8. Central Research Labs., Inc., Report under contract NORD-6119-Task 601, "Summary of Investigations Completed on the Mixed Fungicides in Various Organic Coating Systems, Jan. 1946
9. Hopf, P.P., Paint Manuf. 23:49 (1953)
10. Shapiro, S., Offic. Dig. 30:414 (1958)

Security Classification		DOCUMENT CONTROL DATA - R & D	
<i>(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)</i>			
1. ORIGINATING ACTIVITY (Corporate author) Naval Research Laboratory Washington, D.C. 20390		2a. REPORT SECURITY CLASSIFICATION Unclassified	
		2b. GROUP	
3. REPORT TITLE  FUNGUS-INHIBITIVE COATINGS IN A JUNGLE ENVIRONMENT			
4. DESCRIPTIVE NOTES (Type of report and inclusive dates) Final report on the problem. Unless otherwise notified, this problem will be considered closed 30 days after the issuance of this report.			
5. AUTHOR(S) (First name, middle initial, last name) F.T. Brannan, J.D. Bultman, and J.M. Leonard			
6. REPORT DATE February 19, 1969		7a. TOTAL NO. OF PAGES 16	7b. NO. OF RLFS 10
6a. CONTRACT OR GRANT NO. NRL Problem C03-02		9a. ORIGINATOR'S REPORT NUMBER(S) NRL Report 6849	
b. PROJECT NO. RR007-08-44-5504		9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)	
c.			
d.			
10. DISTRIBUTION STATEMENT  This document has been approved for public release and sale; its distribution is unlimited.			
11. SUPPLEMENTARY NOTES		12. SPONSORING MILITARY ACTIVITY Department of the Navy (Office of Naval Research) Washington, D.C. 20360	
13. ABSTRACT  The ability of ten candidate fungicides, singly and in combination, to suppress fungal growth on a variety of organic coatings under natural, tropical conditions was investigated. After 1 year of exposure, substantial protection for the susceptible coatings was afforded by copper-8-quinolate and binary mixtures of p-toluenesulfonamide with copper-8-quinolate and phenylmercuric phthalate in fairly low concentration. At a higher concentration, p-toluenesulfonamide provided good protection for the susceptible coatings, while its N-ethyl derivative was less effective and its N-cyclohexyl derivative was almost inert. At an intermediate concentration, 2,3-dichloro-1,4-naphthoquinone and p-chlorophenoxyacetic acid provided effective protection for the susceptible coatings. None of the eight binary mixtures tested showed any evidence of synergism. The results confirm those obtained from earlier exposure programs, which showed that p-toluenesulfonamide is effective at higher concentrations in providing protection against fungal infestations. It was observed, however, that at very low levels this material may act as a growth promoter.			

